TRITIUM CONCENTRATION IN ATMOSPHERIC WATER VAPOR AND VEGETATION ADJACENT TO A SPENT NUCLEAR FUEL REPROCESSING PLANT IN JAPAN

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The spent nuclear fuel reprocessing plant located in Rokkasho, Aomori, Japan (40o57'66"N, 141o21'87"E) is in going operation tests with actual spent nuclear fuels, and releases a limited amount of tritium into the surrounding environment. We have measured atmospheric tritium concentrations at pine stands, which locate about 2 km away from the plant, for studying the effects of the release on the environmental tritium level.

Water vapor in outdoor air was collected in the vessel which had a porous polyethylene membrane on the top and contained 500 g of molecular sieves 3A (MS-3A) inside. Water vapor permeated through the membrane was adsorbed by the MS-3A in the vessel.

Fresh pine needles were collected in the pine stands located at Rokkasho. The forest floor is covered with dead pine needles (litter). Samples of about 500 g of the fresh pine needles were collected at these points almost monthly. The tritium concentration in the organic fraction of pine needles collected from in ordinary environment is similar to that of the water fraction. Tritium of the water fraction of plants is called FWT (Free Water Tritium) and that of the organic fraction is called OBT. The FWT is obtained by freeze drying of the sample and the OBT by the combustion of the dried sample.

The pine needles or humus were placed in a desiccator to recover the FWT in the sample by freeze-drying under reduced pressure. The water in the sample was collected in a glass trap cooled at liquid nitrogen temperature installed between the plastic desiccator and the vacuum freeze-drying instrument. This water is described as the FWT water.

The samples were cryogenically processed to separate free water and dried residue. A portion of the residue sample was sealed in an evacuated Al-Si glass ampoule (8252, SCHOTT). To remove atmospheric He as completely as possible to lower background of ^{3}He from ^{3}H, a small amount of acetone was added to the sample, and the ampoule was evacuated to expel adsorbed gases in the sample before sealed. The ^{3}He was measured with a NG-MS (VG-5400, Micromass) after the sealed ampoule was stored at -30oC over 90 d.

The HTO concentrations in air in Rokkasho between November 2007 and December 2009 ranged from 0.3 to 2.2 Bq L^{-1}. The FWT concentrations in the fresh pine needles in Rokkasho between October 2007 and December 2009 ranged from 0.2 to 7.9 Bq L^{-1}. The OBT concentrations in the fresh pine needles in Rokkasho at 2002 ranged from 0.3 to 1.0 Bq L^{-1}. The ^{3}H concentration in the FWT of the fresh pine needles and humus showed a poor correlation with that in water vapor in air. The ^{3}H concentration in the FWT of the fresh pine needles showed a correlation with that in OBT water fraction.

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Performance of the passive sampler was evaluated from the weight of water adsorbed by the MS-3A and absolute humidity in the collecting period. The results showed good adsorption efficiency for atmospheric water vapor of the newly developed sampler. Tritium concentration in water recovered from the MS-3A in the vessel was also compared with that obtained by active sampling method. Both tritium concentrations agreed well with each other, which showed that this method was practical and effective.

We developed an analytical method for OBT in environmental samples by measuring triogenic ^{3}He with a noble gas mass spectrometer, and successfully applied to low-level OBT in samples collected from the current general environment

The FWT and OBT concentrations of the fresh pine needles and humus samples in Rokkasho are presented in Fig. 2.

In these tritium levels, it is difficult to determine the tritium concentration by direct LSC measurement.